

(19)



Europäisches Patentamt

European Patent Office

Office européen des brevets



(11)

EP 0 963 787 A1

(12)

# EUROPEAN PATENT APPLICATION

published in accordance with Art. 158(3) EPC

(43) Date of publication:

15.12.1999 Bulletin 1999/50

(51) Int. Cl.<sup>6</sup>: B01J 13/00, B01F 3/08

(21) Application number: 96939323.0

(86) International application number:

PCT/JP96/03492

(22) Date of filing: 28.11.1996

(87) International publication number:

WO 97/30783 (28.08.1997 Gazette 1997/37)

(84) Designated Contracting States:

DE FR GB

(30) Priority: 20.02.1996 JP 3188296

(71) Applicant:

JAPAN as represented by DIRECTOR GENERAL  
OF NATIONAL FOOD  
RESEARCH INSTITUTE, MINISTRY OF  
AGRICULTURE, FORESTRY AND FISHERIES  
Tsukuba-shi, Ibaraki 305 (JP)

• KIKUCHI, Yuji

Ryugasaki-shi, Ibaraki 301 (JP)

• KAWAKATSU, Takahiro,

Room 203,

Mezondo.Byu

Sendai-shi, Miyagi 982 (JP)

(74) Representative:

Jackson, Robert Patrick et al

Frank B. Dehn & Co.,

European Patent Attorneys,

179 Queen Victoria Street

London EC4V 4EL (GB)

(72) Inventors:

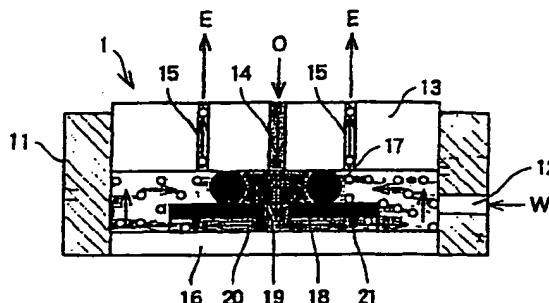
• NAKAJIMA, Mitsutoshi

Tsukuba-shi, Ibaraki 305 (JP)

## (54) METHOD AND DEVICE FOR PRODUCING EMULSIONS

(57) A dispersed phase (O) supplied inside a bulk-head member (17) through a supply port (14) enters a gap (20) between a plate (16) and a base (18) through a supply port (19) of the base (18) and the dispersed phase (O) having entered the gap (20) then enters a continuous phase (W) through a boundary section (21) by virtue of pressure applied by a pressurizing means such as a pump. Then, the dispersed phase is made into a particle having a predetermined diameter by a microchannel (24) in passing through this boundary section (21), thereby forming an emulsion (E) in which the dispersed phase (O) of the predetermined diameter is dispersed in the continuous phase (W).

FIG.2



BEST AVAILABLE COPY

## Description

### Technical Field

[0001] The present invention relates to a method of producing emulsions used in the food industry, manufacturing of drugs and cosmetics, etc. and to a device implementing the method.

### Background Art

[0002] Techniques in which a biphasic system for which a separated state is thermodynamically stable, such as that composed of a water phase and an organic phase, is emulsified to obtain a semi-stable emulsion have been conventionally known. For general emulsification methods, as described in "Science of Emulsions" (Asakura-shoten, 1971), methods using a mixer, a colloid mill, a homogenizer, etc. and a method of dispersion with a sound wave are known.

[0003] These general methods mentioned above have such a disadvantage that diameters of dispersed phase particles in a continuous phase distribute over a wide range.

[0004] Then, a method using filtration through a membrane comprising polycarbonate (Biochemica et Biophysica Acta, 557 (1979), North-Holland Biochemical Press); a method using repeated filtrations through a PTFE (polytetrafluoroethylene) membrane (Proceedings of the 26th Autumn Meeting of the Society of Chemical Engineers, Japan, 1993); and a method of manufacturing homogenous emulsions by sending a dispersed phase into a continuous phase through a porous glass membrane with uniform pores (Japanese Patent Application Laid-Open No. 2-95433) have been proposed.

[0005] As a method of producing emulsions using a nozzle or a porous plate, a laminar-flow dripping method (KAGAKU K OGAKU vol. 21, No. 4, 1957) is also known.

[0006] Among the conventional modified methods mentioned above, the method using filtration through a membrane comprising polycarbonate and the method using repeated filtrations through a PTFE membrane cannot manufacture emulsions of particles larger than membrane pores theoretically or cannot separate particles smaller than membrane pores. These methods are therefore especially unsuitable for producing emulsions of large particles.

[0007] In the method using a porous glass membrane with uniform pores, as shown in a graph in Figure 11, when an average diameter of membrane pores is small, particle diameters distribute in a narrow range and thus homogenous emulsions can be obtained. When an average diameter of membrane pores is increased, however, particle diameters distribute over a wide range so that homogenous emulsions cannot be obtained.

[0008] In addition, in the laminar-flow dripping

method, particle sizes become 1,000  $\mu\text{m}$  or more and distribute over a wide range so that homogenous emulsions cannot be obtained.

### Disclosure of the Invention

[0009] The present invention aims at solving the above-mentioned problems of prior art and intends to provide a method and a device for efficiently producing homogenous emulsions without broadening a distribution of particle diameters.

[0010] In order to achieve this object, according to the method for producing emulsions of the present invention, a pressurized dispersed phase is forced to be sent into a continuous phase through a large number of microchannels with a predetermined width formed on a base, etc.

[0011] In the device for producing emulsions of the present invention, a base on which a supply port for a dispersed phase is formed is provided, a gap to which the dispersed phase is supplied is formed between the base and a plate placed opposing the base, a boundary section between the dispersed phase and a continuous phase is formed on a side opposing the plate of the base, a large number of microchannels with a predetermined width are formed in the boundary section, and the dispersed phase is brought into contact with the continuous phase via the microchannel.

[0012] The plate opposing the base may be a transparent plate. It enables direct optical observation of channel moving of a dispersed phase and a condition of contact between the dispersed phase and a continuous phase, thereby enabling to control production of emulsions.

[0013] The boundary section, in which a large number of microchannels are formed, is provided so as to surround the supply port for the dispersed phase so that emulsions can be produced efficiently.

[0014] As a method of forming microchannels, a large number of microchannels with a predetermined width can be formed in any shape on the base by applying an etching process, one of the manufacturing steps of semiconductor integrated circuits.

### Brief Description of the Drawings

[0015]

Figure 1 shows an entire system incorporating the device for producing emulsions according to the present invention;

Figure 2 is a vertical section of the device for producing emulsions;

Figure 3 is an exploded perspective view of the necessary parts of the device for producing emulsions;

Figure 4 is a bottom view of a base constituting the device for producing emulsions;

Figure 5 is a top view of a base constituting the

device for producing emulsions;

Figure 6 is an enlarged perspective view of a boundary section formed on the base;

Figure 7 shows microchannels;

Figure 8 is a cross sectional view of another example of the device for producing emulsions;

Figures 9 (a)- (d) specifically show fine patterns formed on the base of Example 1 and Figure 9 (e) shows colloids of Example 1;

Figure 10 specifically shows colloids of Example 2; and

Figure 11 is a graph showing the relation between average membrane pore diameters and distribution of particle diameters of emulsions in using a conventional device.

#### Description of Symbols

[0016] In these figures, 1 denotes a device for producing emulsions, 2 a dispersed phase tank, 5 a continuous phase tank, 9 an emulsion tank, 11 a body of a device for producing emulsions, 12 a supply port for a continuous phase (W), 14 a supply port for a dispersed phase (O), 15 a collection port for emulsion (E), 16 a plate, 17 a bulkhead member, 18 a base, 19 a supply port for a dispersed phase (O), 20 a gap, 21 a boundary section, 22 a protruded bar, 23 a protrusion, 24 a microchannel, 25 an optical reading device, O a dispersed phase, W a continuous phase, and E emulsion.

#### Best Mode for Carrying Out the Invention

[0017] An embodiment of the present invention is now described referring to attached figures. Figure 1 shows an entire system incorporating the device of producing emulsions according to the present invention. A dispersed phase (O) is supplied from a dispersed phase tank 2 through a pump 3 and a piping 4 to a device for producing emulsions 1 and a continuous phase (W) is supplied from a continuous phase tank 5 through a pump 6 and a piping 7 to the device for producing emulsions 1. An emulsion (E) produced in the device for producing emulsions 1 is stored in an emulsion tank 9 through piping 8.

[0018] A pump 6 is shown as a pressurizing means in this example, but other pressurizing means such as a difference in water level can be used.

[0019] Next, a structure of the device for producing emulsions 1 is explained referring to Figure 2 to Figure 7. Here, Figure 2 is a vertical section of the device for producing emulsions, Figure 3 is an exploded perspective view of necessary parts of the device for producing emulsions, Figure 4 is a bottom view of a base constituting the device for producing emulsions, Figure 5 is a top view of a base constituting the device for producing emulsions, Figure 6 is an enlarged perspective view of a boundary section formed on the base, and Figure 7 shows microchannels.

[0020] In the device for producing emulsions 1, the supply port 12 for a continuous phase (W) with which the piping 7 connects is formed on a side wall of a body 11, an upper opening of the body 11 is closed with a lid 13, a supply port 14 for a dispersed phase (O) with which the piping 4 connects is formed on the center of the lid 13, and a collection port 15 for emulsion (E) which is connected with the piping 8 is formed at a place apart from the center of the lid 13.

[0021] A lower opening of the body 11 is closed with a plate 16 and a base 18 is placed inside the body 11 with a bulkhead member 17 such as an O-ring lying between the base and the lid 13. The bulkhead member 17 separates the supply port 14 for the dispersed phase (O) from the collection port 15 for emulsion (E).

[0022] In addition, a supply port 19 for a dispersed phase (O) is formed in a center part of the base 18, a gap 20 is formed between the base 18 and the plate 16 placed opposing the base 18, and a boundary section 21 between a dispersed phase (O) and a continuous phase (W) is formed on a side opposing the plate 16 of the base 18.

[0023] In the boundary section 21, a protruded bar 22 is formed on the periphery of the base so as to surround the supply port 19 for a dispersed phase (O) rectangularly, a large number of protrusions 23 are formed on the protruded bar 22 with predetermined intervals, and spaces between the protrusions 23, 23 are used as microchannels 24.

[0024] The boundary 21 section with these microchannels 24 is formed by wet or dry etching.

[0025] In the above, a dispersed phase (O) supplied inside the bulkhead member 17 through the supply port 14 enters a gap 20 between the plate 16 and the base 18 through the supply port 19 of the base 18 and the dispersed phase (O) having entered the gap 20 enters a continuous phase (W) through the boundary section 21 by virtue of pressure applied by a pressurizing means such as a pump. Then, the dispersed phase is made into a particle having a predetermined diameter by a microchannel 24 in passing through the boundary section 21, thereby forming an emulsion (E) in which the dispersed phase (O) of the predetermined diameter is dispersed in the continuous phase (W).

[0026] The plate 16 may be a transparent plate such as a glass plate. The use of a transparent plate enables observation from the outside using the optical reading device 25 as shown in Figure 3 and allows precise control of a rate of producing emulsions accompanying a change in driving pressure.

[0027] In addition, a type of emulsion produced can be changed according to a hydrophilic or hydrophobic property of a base. That is, when a hydrophilic base is used, an O/W type emulsion can be produced, and when a hydrophobic base is used, a W/O type emulsion can be produced.

[0028] In addition, a constitution of a device for producing emulsions 1 is not restricted to the one

described above. For example, as shown in Figure 8, a device for producing emulsions can have such a structure that a supply port 14 for a dispersed phase (O) is formed on an edge part close to one side of the body 11 of a lid 13 and a collection port 15 for emulsions (E) is formed in almost the center of the lid 13. It is also possible to form a channel in which a dispersed phase (O) flows by sticking two bases in a manner that the sides on which microchannels are formed are opposed.

[0029] Specific examples will be described below.

(Example 1)

[0030] An emulsion was attempted to be produced using triolein containing 0.3wt% sorbitan monolaurate as a dispersed phase and water as a continuous phase under driving pressures of 2.65 kPa, 3.97 kPa, 4.41 kPa, and 8.38 kPa. The results are shown in Figures 9 (a)-(d).

[0031] Figures 9 (a) - (c) indicate that when a driving pressure is low such as 2.65 kPa, 3.97 kPa, and 4.41 kPa, it is impossible to force a dispersed phase to enter a continuous phase so that an emulsion is not produced. However, it is shown that when a driving pressure is elevated to 8.38 kPa, a dispersed phase is sent into a continuous phase through microchannels as shown in Figure 9 (d). It is observed that the emulsion thus produced is homogenous with a very uniform particle diameter as shown in Figure 9(e).

(Example 2)

[0032] An emulsion was attempted to be produced using water containing 0.75wt% KCl as a dispersed phase and kerosene containing 3wt% sorbitan monolaurate as a continuous phase under a driving pressure of 1.62 kPa. The results are shown in Figure 10. As shown in Figure 10, a homogenous emulsion with a markedly uniform particle diameter is observed.

[0033] As described above, according to the method of producing emulsions of the present invention, since pressurized dispersed phase is forced to be sent into a continuous phase through a large number of microchannels with a predetermined width formed on a base or the like, a distribution of particle diameter does not broaden and homogenous emulsions can be obtained, when dispersed particles have a small diameter, as a matter of course, and even when dispersed particles have a larger diameter.

[0034] According to the device for producing emulsions of the present invention, since a base on which a supply port for a dispersed phase is formed is provided, a gap to which the dispersed phase is supplied is formed between the base and a plate placed opposing the base, a boundary section between the dispersed phase and a continuous phase is formed on a side opposing the plate of the base, a large number of microchannels with a predetermined width are formed in the

boundary section, and the dispersed phase is brought into contact with the continuous phase via the microchannels, a distribution of particle diameter does not broaden and homogenous emulsions can be efficiently produced.

[0035] The use of a transparent plate for a plate opposing a base enables direct optical observation of channel moving of a dispersed phase and a condition of contact between the dispersed phase and a continuous phase, thereby enabling to control production of emulsions

[0036] The provision of a boundary section, in which a large number of microchannels are formed, so as to surround the supply port for a dispersed phase allows further efficient emulsion production.

[0037] The adoption of an etching method for forming microchannels allows accurate formation of microchannels in any shape with a predetermined width.

#### Industrial Applicability

[0038] The method and device for producing emulsions according to the present invention can contribute to production of emulsions utilized in the food industry and manufacturing of drugs and cosmetics, etc.

#### Claims

1. A method for producing emulsions wherein a pressurized dispersed phase is forced to be sent into a continuous phase through a large number of microchannels with a predetermined width.
2. A device for producing emulsions by sending a dispersed phase into a continuous phase wherein a base on which a supply port for a dispersed phase is formed is provided, a gap to which the dispersed phase is supplied is formed between the base and a plate placed opposing the base, a boundary section between the dispersed phase and a continuous phase is formed on a side opposing the plate of said base, a large number of microchannels with a predetermined width are formed in the boundary section, and the dispersed phase is brought into contact with the continuous phase via the microchannels.
3. A device for producing emulsions according to claim 2 wherein a plate opposing said base is a transparent plate.
4. A device for producing emulsions according to claim 2 wherein the boundary section in which a large number of said microchannels are formed is provided so as to surround a supply port for a dispersed phase.
5. A device for producing emulsions according to any

one of claims 2-4 wherein said microchannels are formed by subjecting the base to precise processing technique such as etching treatment, irradiation of electron rays, or CVD method.

5

10

15

20

25

30

35

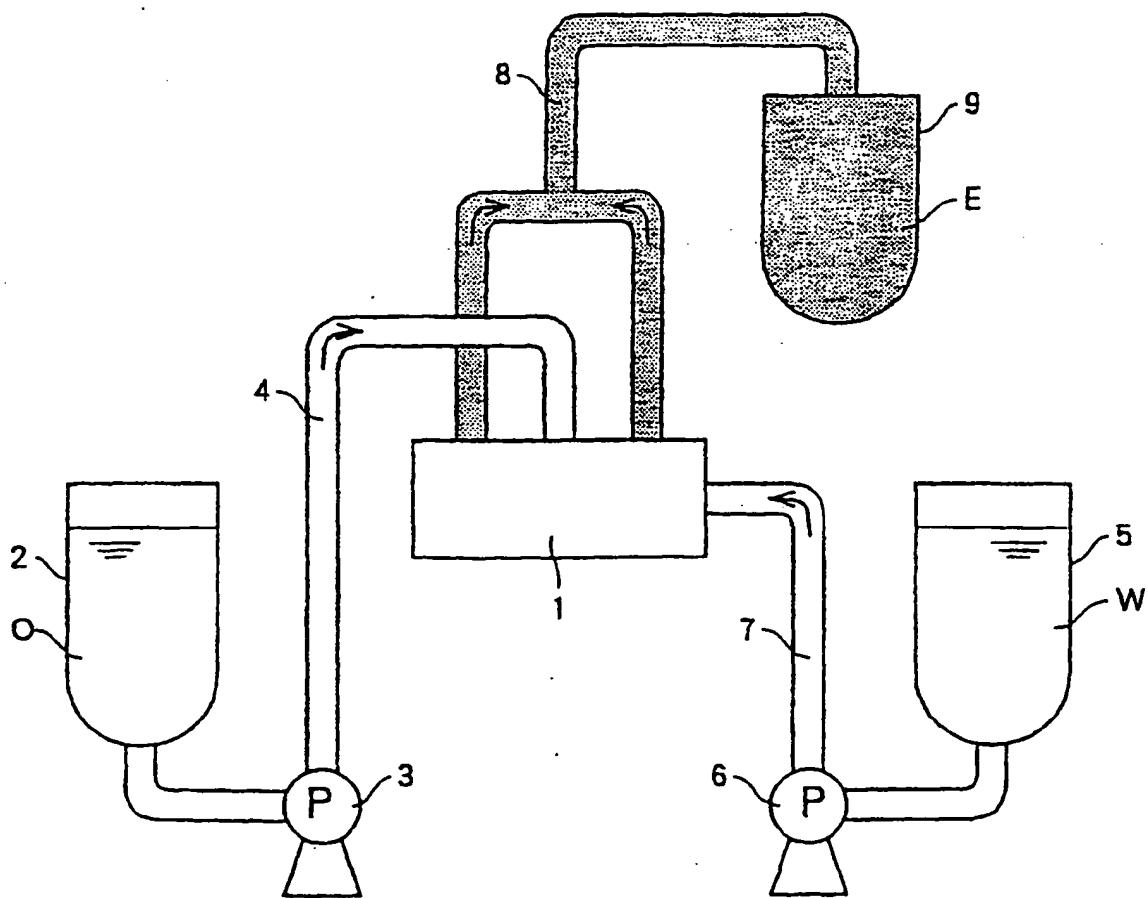
40

45

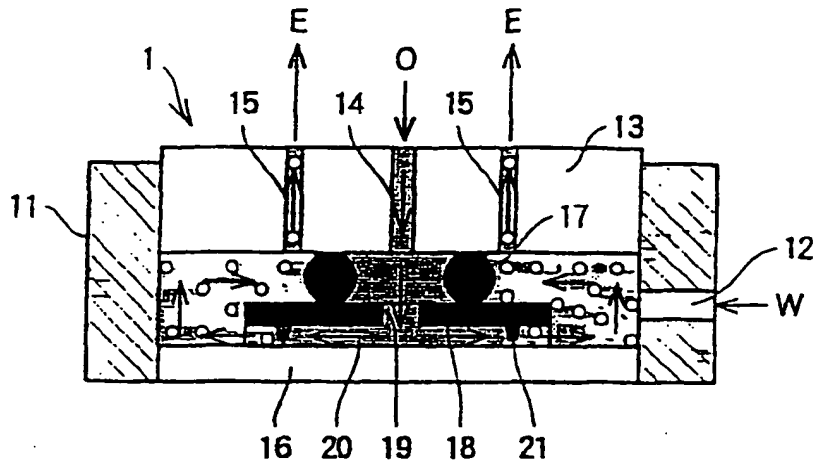
50

55

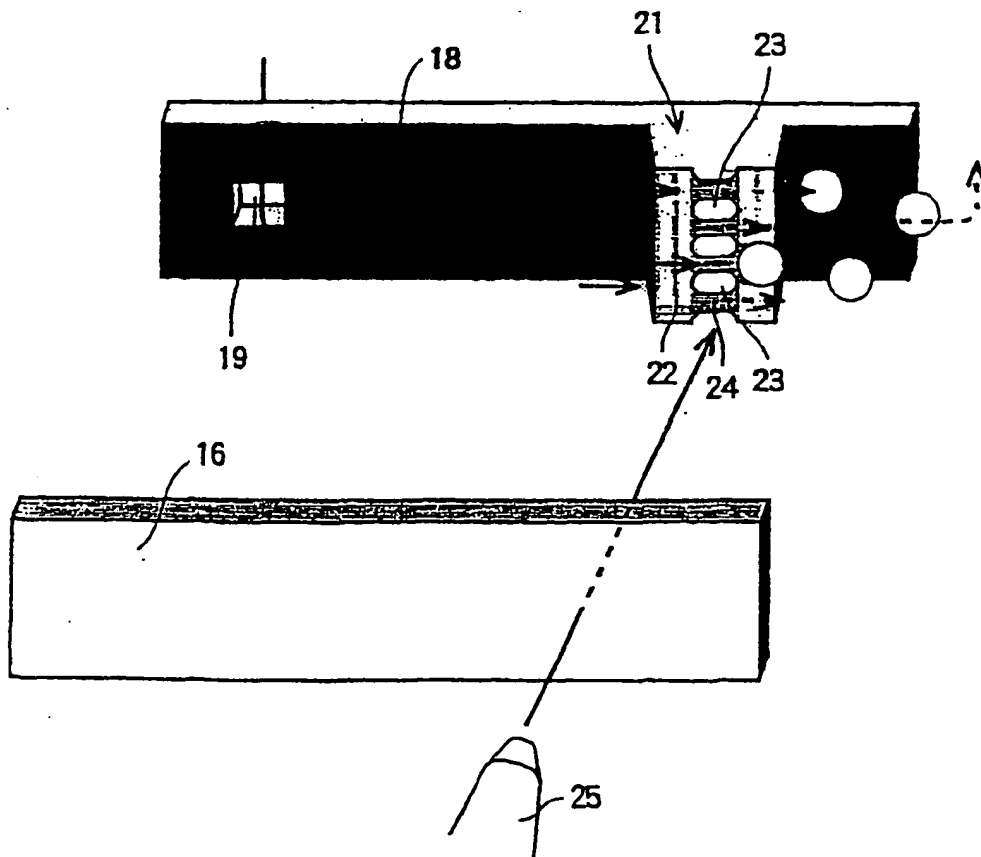
**FIG.1**



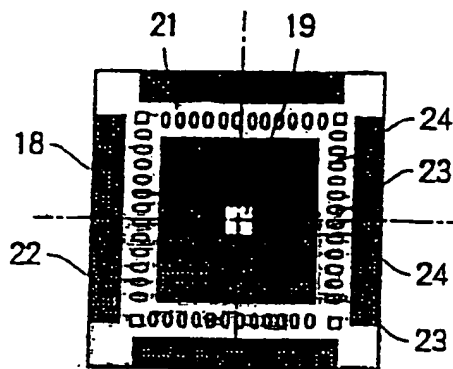
**FIG. 2**



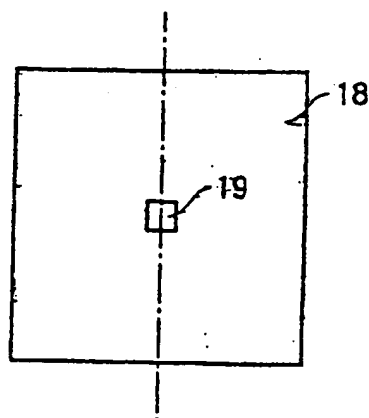
**FIG. 3**



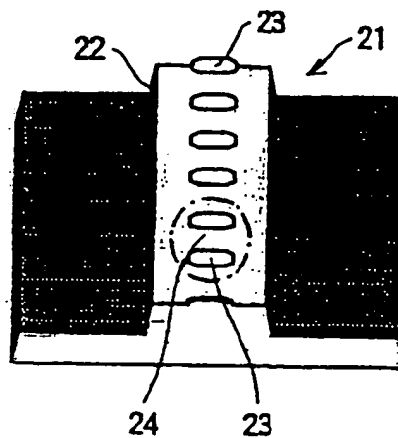
**FIG. 4**



**FIG. 5**

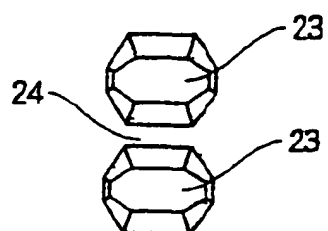


**FIG. 6**

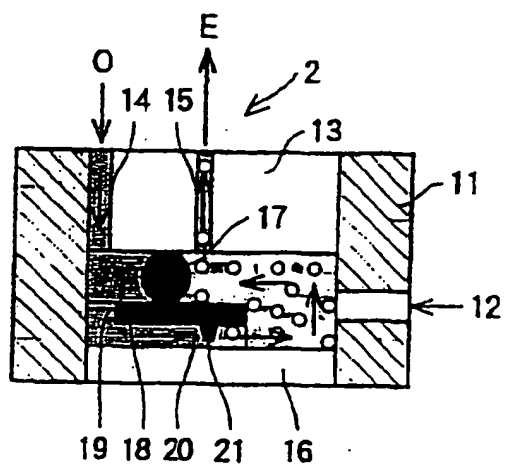


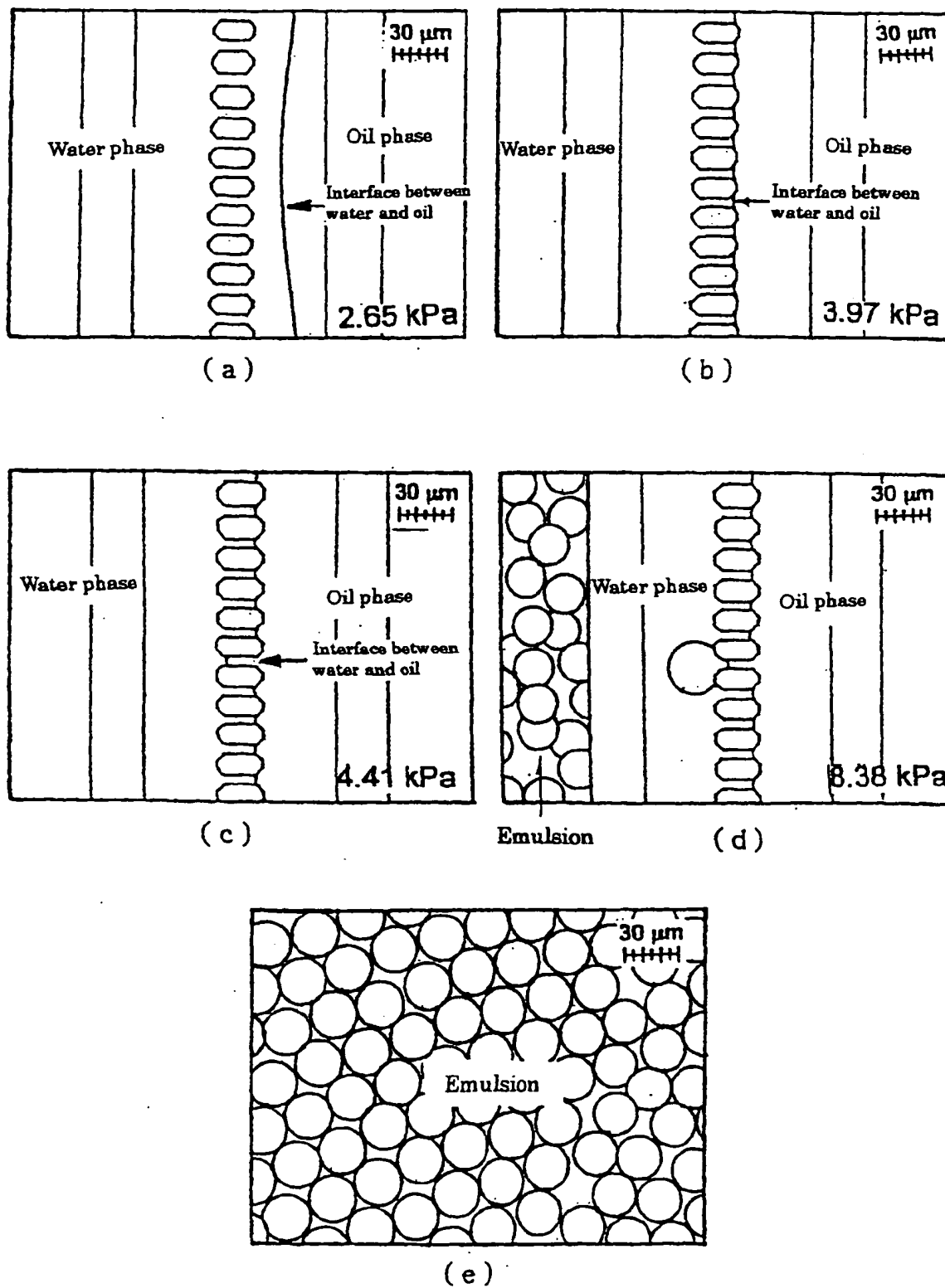


**FIG. 7**

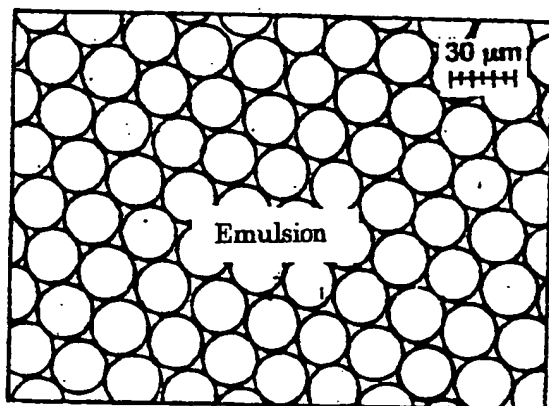


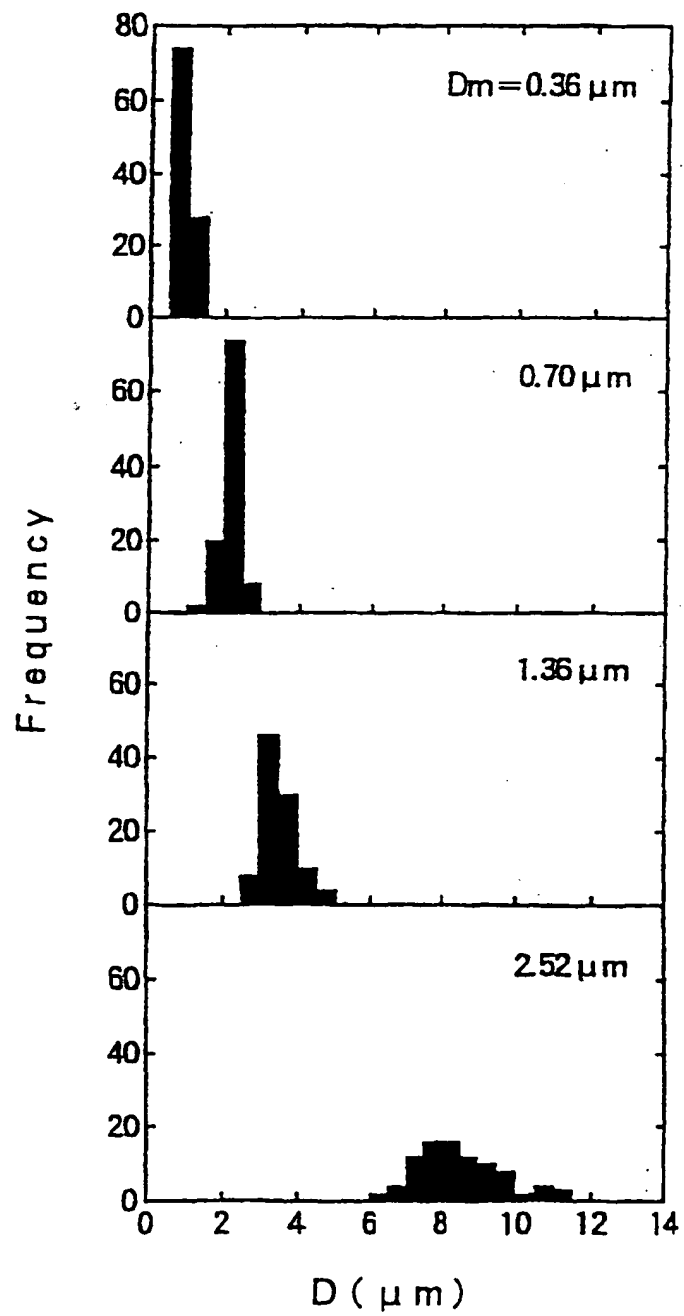
**FIG. 8**



**FIG. 9**

**FIG.10**



**FIG. 11**

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP96/03492

## A. CLASSIFICATION OF SUBJECT MATTER

Int. Cl<sup>6</sup> B01J13/00, B01F3/08

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int. Cl<sup>6</sup> B01J13/00, B01F3/08, 5/06

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1926 - 1996	Jitsuyo Shinan Keisai
Kokai Jitsuyo Shinan Koho	1971 - 1996	Koho
Toroku Jitsuyo Shinan Koho	1994 - 1996	1996

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP, 5-220382, A (Miyazaki Prefecture), August 31, 1993 (31. 08. 93), Claim 1 & WO, 9300156, A1 & EP, 546174, A1 & US, 5326484, A	1
X	JP, 6-315617, A (Miyazaki Prefecture), November 15, 1994 (15. 11. 94), Claim 1 (Family: none)	1
Y	JP, 6-71150, A (Nomura Micro Science Co., Ltd.), March 15, 1994 (15. 03. 94), Claim 2; Fig. 1 (Family: none)	2, 5
A	JP, 60-5223, A (Paul Corp.), January 11, 1985 (11. 01. 85), Claim 2 & GB, 2141037, A & EP, 130681, A & FR, 2547212, A & AU, 8429080, A & NO, 8402281, A & ZA, 8404276, A & PT, 78694, A & BR, 8402659, A & US, 4534388, A & CA, 1215958, A & CH, 659779, A & GB, 2141037, B	4

☒ Further documents are listed in the continuation of Box C.☐ See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&amp;" document member of the same patent family

Date of the actual completion of the international search

December 18, 1996 (18. 12. 96)

Date of mailing of the international search report

January 8, 1996 (08. 01. 96)

Name and mailing address of the ISA/

Japanese Patent Office

Authorized officer

Facsimile No.

Telephone No.

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP96/03492

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	& EP, 130681, B & DE, 3481175, G JP, 54-116389, A (Exxon Research and Engineering Co.), September 10, 1979 (10. 09. 79), Claim 1 & NL, 7900345, A & SE, 7900350, A & DE, 2901501, A & US, 4201691, A & CH, 641974, A & IT, 1114701, B	1

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☐ FADED TEXT OR DRAWING
- ☐ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☐ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☒ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**